PB-220 708

OBSERVATION OF AIRBORNE TRITIUM WASTE DISCHARGE FROM A NUCLEAR FUEL REPROCESSING PLANT

Joseph A. Cochran, et al

Office of Radiation Programs
Washington, D.C.

February 1973

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Office of Radiation Programs

34

BIBLIOGRAPHIC DATA SHEET	1. Report No. EPA/ORP 73-1	2.	3. Recipient's Accession No. PB-220 708
4. Title and Subtitle OBSERVATIONS OF AIRBORNE TRITIUM WASTE DISCHARGE FROM A NUCLEAR FUEL REPROCESSING PLANT			5. Report Date (Issue) February 1973 6.
7. Author(s) Joseph A. Cochran, William R. Griffin, Jr., Emilio J. Troinello			8. Performing Organization Rept. No.
9. Performing Organization Name and Address U.S. ENVIRONMENTAL PROTECTION AGENCY Office of Radiation Programs Washington, D.C. 20460			10. Project/Task/Work Unit No. 11. Contract/Grant No.
12. Sponsoring Organization Name and Address See item 9.			13. Type of Report & Period Covered Final 14.

15. Supplementary Notes

16. Abstracts

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17. Key Words and Document Analysis. 17a. Descriptors

Fuel Reprocessing, Environment, Nuclear Facility, Population Dose, Sampling, Tritium, Waste Discharge

17b. Identifiers/Open-Ended Terms

17c. COSATI Field/Group 18E, 18G, 18H

18. Availability Statement	19. Security Class (This Report)	21. No. of Pages
Release unlimited	UNCLASSIFIED	40 34
	20. Security Class (This Page) UNCLASSIFIED	22. Price $3.00 $1.45

NTIS-35 (REV. 3-72) THIS FORM MAY BE REPRODUCED USCOMM-DC 14952-P72

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Joseph A. Cochran
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Emilio J. Troianello

February 1973

U.S. ENVIRONMENTAL PROTECTION AGENCY
Office of Radiation Programs
Field Operations Division
Washington, D.C. 20460

FOREWORD

The Office of Radiation Programs carries out a national program designed to evaluate the exposure of man to ionizing and nonionizing radiation, and to promote development of controls necessary to protect the public health and safety and assure environmental quality.

Within the Office of Radiation Programs, the Field Operations Division conducts programs relating to sources and levels of environmental radioactivity and the resulting population radiation dose. Reports of the findings are published in the monthly publication, *Radiation Data and Reports*, appropriate scientific journals, and Division technical reports.

The technical reports of the Field Operations Division allow comprehensive and rapid publishing of the results of intramural and contract projects. The reports are distributed to State and local radiological health programs, Office of Radiation Programs technical and advisory committees, universities, libraries and information services, industry, hospitals, laboratories, schools, the press, and other interested groups and individuals. These reports are also included in the collections of the Library of Congress and the National Technical Information Service.

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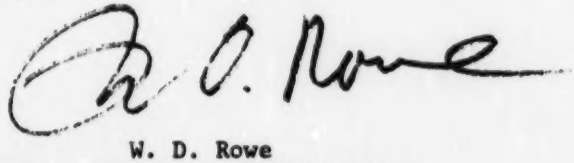

W. D. Rowe
Deputy Assistant Administrator
for Radiation Programs

PREFACE

The Office of Radiation Programs of the Environmental Protection Agency, in cooperation with the New York State Department of Environmental Conservation, Nuclear Fuel Services, Inc., and the Atomic Energy Commission, have, over the past several years, conducted studies at the Nuclear Fuel Services, the nation's first commercial nuclear fuel reprocessing plant. The overall purpose of this study was to determine the requirements of an environmental surveillance program for fuel reprocessing plants. Specific study objectives included (a) characterization of both the gaseous and liquid waste effluents from the plant, (b) measurement of the environmental concentrations of the discharged radionuclides, and (c) delineation of the critical exposure pathways and estimation of the radiation doses to the population living near the plant. Earlier results of this study were published in a series of reports:

(1) BRH/NERHL 70-1 *An Estimate of Radiation Doses Received by Individuals Living in the Vicinity of a Nuclear Reprocessing Plant*

(2) BRH/NERHL 70-2 *Liquid Waste Effluents from a Nuclear Fuel Reprocessing Plant*

(3) BRH/NERHL 70-3 *An Investigation of Airborne Radioactive Effluent from an Operating Nuclear Fuel Reprocessing Plant*

(4) BRH/NERHL 70-4 *Calibration and Initial Field Testing ^{85}Kr Detectors for Environmental Monitoring*

(5) ORP/SID 72-5 *Iodine-129 in the Environment Around a Nuclear Fuel Reprocessing Plant*

This report presents the results of followup studies on the tritium discharges from Nuclear Fuel Services, on the concentrations of this radionuclide in the environment around the plant, and resultant doses to the population in the vicinity of the plant.

Charles L. Weaver

Charles L. Weaver
Director
Field Operations Division

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ACKNOWLEDGMENTS

The assistance and cooperation of the following individuals in planning and carrying out this study is gratefully acknowledged.

John Russell
Office of Radiation Programs
Environmental Protection Agency
Floyd Galpin
Office of Radiation Programs
Environmental Protection Agency
Charles Weaver
Office of Radiation Programs
Environmental Protection Agency
Thomas Cashman
Bureau of Radiological Pollution Control
New York State Department of Environmental Conservation
Gene Michaels
Bureau of Radiological Pollution Control
New York State Department of Environmental Conservation
Robert Wozniak
Bureau of Radiological Pollution Control
New York State Department of Environmental Conservation
Edward North
Nuclear Fuel Services
Terry Wenstrand
Nuclear Fuel Services
Dave Wilcox
Nuclear Fuel Services

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ABSTRACT

A study was conducted at Nuclear Fuel Services, Inc. (NFS) to: (a) characterize the stack tritium effluent in the gaseous and water vapor forms, during normal plant operations, (b) determine the weekly average tritium air concentrations at five selected sites around the plant over a 4-month period, (c) evaluate methods used for sampling at the stack and in the environment, and (d) estimate the dose to the population in the immediate vicinity of the plant from tritium stack effluents.

Characterization of tritium release from the stack was carried out during three reprocessing campaigns assumed to be typical of the NFS operation. Evaluation of the data indicates that the tritium release rate from the stack is 6.75×10^{-4} curies per megawatt-day burnup. Approximately 25 percent of the activity is in the gaseous state and 75 percent is in the water vapor state. The average annual release of tritium via the stack, considering the total fuel processed from 1966 to 1971, is estimated to be 500 curies per year. Data for the five air sampling stations around the site indicate that the plant contribution to the atmosphere is small.

OBSERVATION OF AIRBORNE TRITIUM WASTE DISCHARGE FROM A NUCLEAR FUEL REPROCESSING PLANT

INTRODUCTION

During the reprocessing of nuclear fuel, tritium, a fission product originating in the fuel elements, is released to the environment (1). The tritium may be released in the gaseous effluent via the stack and/or in the low-level liquid wastes. If discharged principally to the atmosphere, tritium is expected to be a predominant contributor to the local population whole body radiation dose ascribable to a fuel reprocessing plant. The objectives of the present study were to:

(a) determine the nature of tritium released from a fuel reprocessing plant,
(b) understand the relation of this release to population dose, and
(c) develop specific methods for detecting tritium onsite and in the environment around a fuel reprocessing plant.

There have been many studies dealing with the sources of tritium and transport of this nuclide in the environment (1-3). These studies have added valuable information to the general subject of tritium impact in the environment. Studies to date at Nuclear Fuel Services, Inc. (NFS), a fuel reprocessing plant located at West Valley, N.Y., have derived information concerning tritium in liquid waste discharge from the plant (4) and levels of airborne tritiated water vapor concentration (activity/volume of water) in the environment surrounding the plant (5). In addition to these studies, some preliminary work was performed to determine the amount of tritiated water vapor discharged through the NFS stack during the fuel dissolution cycle (6).

The present study was undertaken at NFS to supplement the studies cited above.

SAMPLING DESCRIPTION

In-plant sampling operations

In-plant sampling was carried out to characterize the tritium stack effluent. A sampler was designed to selectively collect the water vapor and the gaseous components of tritium from the stack effluent. The tritium sampler was operated in parallel with the NFS stack monitoring system which allowed isokinetic sampling at the 80-foot level of the 200-foot stack (6).

The tritium sampler was a flow-through system consisting of a desiccant which removes water vapor, a catalytic burner which oxidizes hydrogen compounds to water, and a second desiccant which removes the oxidation products in the form of water vapor. Carrier hydrogen was used in the burner stage to provide an adequate volume of combustion products for collection in the second desiccant stage (7).

The first desiccant stage of the system was operated when only a water vapor sample was of interest. The burner and second desiccant were put on-line when either a gaseous tritium sample or simultaneous water vapor and gaseous tritium samples were desired. The sampling train contained taps located after the first desiccant stage which enabled 2.8 liter evacuated tank grab samples of the gaseous tritium components to be obtained. A detailed description of the basic tritium stack sampling system and analytical technique appears in appendix A.

Stack sampling was carried out over a 6-month period from June to November 1971. During this sampling period, the plant processed four separate fuel inventories or campaigns. These campaigns are considered to be representative of the normal type of fuel reprocessing carried out at NFS. A listing of campaigns and associated information on the fuel inventories are given in appendix B.

The specific samples that were obtained are listed below:

(a) Weekly stack water vapor samples were obtained from June 7, 1971 to November 30, 1971. Twenty-five samples were collected in this period. The purpose of this sampling was to obtain weekly stack concentrations for correlation with fuel throughput and offsite tritium air concentrations during the same period.
(b) Simultaneous water vapor and gaseous tritium samples were obtained during the sixth, seventh, and eighth dissolution runs of the Humboldt Bay fuel campaign in order to measure total tritium in-stack during a dissolution cycle.
(c) Simultaneous water vapor and gaseous tritium samples were also obtained during the complete campaigns of the Parr and Big Rock Point fuel inventories. These data were necessary in order to determine the relative tritium concentrations present as gas (HT) and water vapor (HTO) during a complete reprocessing campaign.
(d) Gas samples were obtained during the three Humboldt Bay dissolutions mentioned above, using evacuated tanks. These samples consisted of dried stack gas containing the gaseous tritium component. The purpose of these samples was to cross-check the values obtained from the desiccant samples obtained during the same time period.
(e) A low-level liquid waste composite sample was obtained at the interceptor for the entire Humboldt Bay campaign period. This sample was used with the stack samples obtained during the Humboldt Bay campaign in order to estimate the total tritium inventory during a campaign period.

Field sampling operations

The field tritium air samplers collected water vapor by use of a desiccant and operated similar to the first stage of the stack tritium sampler. A detailed description of the field sampler appears in appendix A.

Five samplers were located around the perimeter of the NFS site. Figure 1 is a map showing the location of each station in terms of azimuth and distance from the plant. The average station distance was approximately 2.6 kilometers (1.6 miles) from the plant whereas the average property line distance is approximately 1.9 kilometers (1.2 miles). A control station, located in Winchester, Mass., was operated on a weekly basis as were the stations around NFS.

Samples were collected weekly from June 7 to October 4, 1972, a total of 17 weeks. During the sampling period, wind-rose data were accumulated in order to determine if any correlation existed between these data and the observed tritium levels at the field stations.

RESULTS AND DISCUSSION

In-plant sample results

Weekly water vapor samples

Tritium water vapor samples were obtained from the stack during the period of June 7 to November 30, 1971. Four campaigns were processed during this period. Figure 2 shows a comparison of the weekly tritium stack concentrations in the vapor form with the weekly plant fuel processing load expressed in terms of burnup.[1] The tritium effluent data were displaced forward in time by 1 week to account for the lag between dissolution of the fuel and the effluent discharge as water vapor. Since the amount of tritium present in the spent fuel is essentially constant when expressed in terms of burnup, the quantity discharged per megawatt-day (MWD) should be relatively constant. This comparison (figure 2) indicates that the quantity of tritium vapor discharged per MWD is not constant and may be dependent on other factors such as fuel cladding, tritium inventory in the process solutions from previous campaigns, and specific processing operations at any given time. However, there is a general correlation where the tritium vapor effluent increases with an increase in the burnup of the fuel being processed.

The tritium water vapor yield for each campaign is given in table 1 in terms of curie per MWD burnup. These values were derived by summing the weekly activity values over the entire campaign period.

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[1]Burnup is an expression of megawatt-days of energy generated by the fuel.

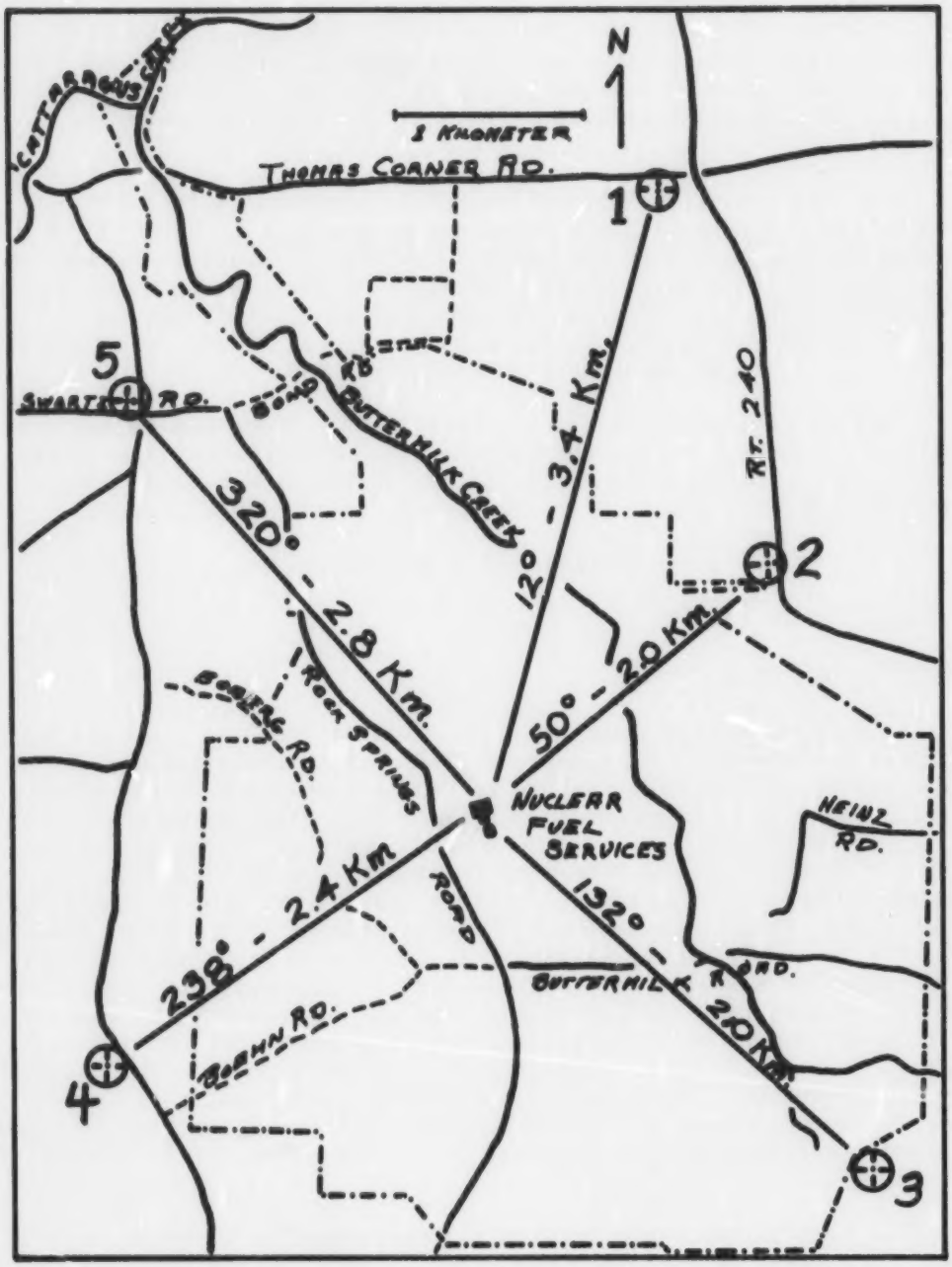

Figure 1. Field tritium sampling locations

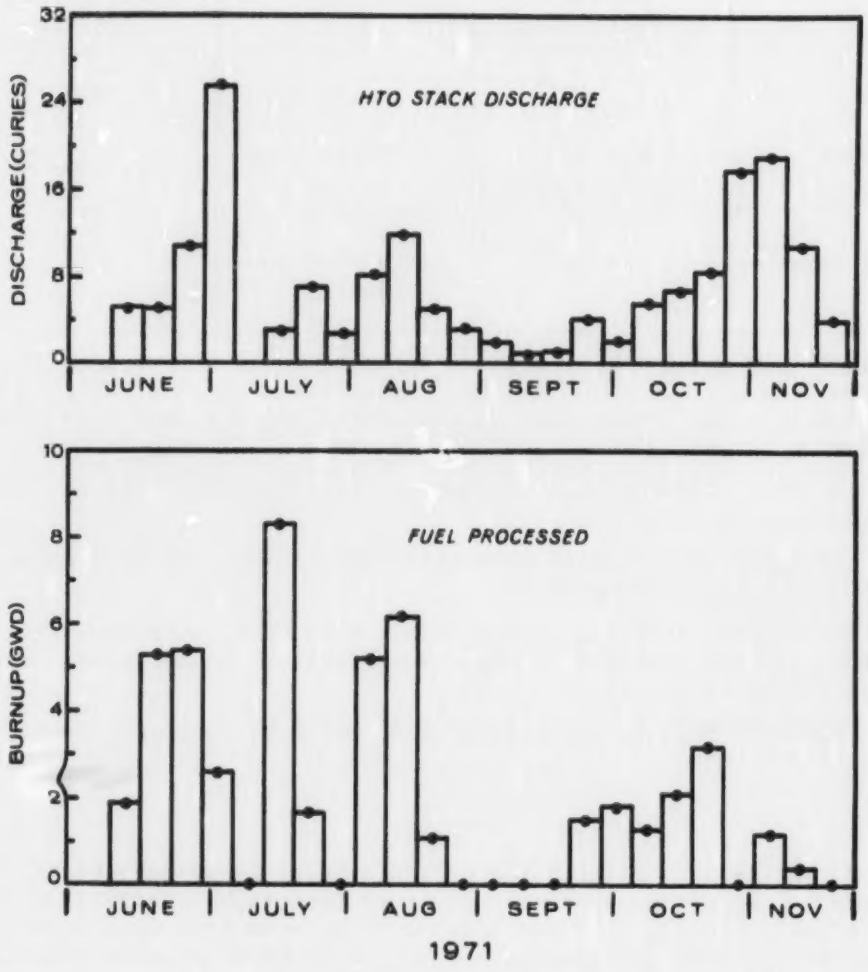

Figure 2. Comparison of weekly tritium stack output with weekly burnup inventory of fuel processed

Table 1. Tritium stack output in the form of water vapor for campaign periods

Campaign	Release per unit burnup[a] (Ci/MW(t)-day)	Cladding material	Fuel age prior to reprocessing (years)
Humboldt Bay[b]----	3.1×10^{-4}	Stainless steel	3.5
Yankee Rowe[c]-----	2.0×10^{-4}	Stainless steel	.8
Parr[d]----------	4.5×10^{-4}	Zircaloy-4	4.8
Big Rock Point[e]--	7.9×10^{-4}	Zircaloy-2 and Iconel	1.6

[a]Based upon weekly tritium water vapor composites throughout the entire campaign periods. The combined analytical and sampling 1σ errors for the weekly samples that made up the composites are less than 12 percent. Burnup, cladding type, and age data from plant records (9).

[b]A 240 MW(t) General Electric Boiling Water Reactor operated by the Pacific Gas and Electric Company.

[c]A 600 MW(t) Westinghouse Pressurized Water Reactor operated by the Yankee Atomic Electric Company.

[d]The Carolinas-Virginia Tube Reactor, a 64 MW(e) pressure tube, heavy water reactor operated by Carolinas-Virginia Nuclear Power Associates, Inc., from 1963 to 1967.

[e]A 240 MW(t) Boiling Water Reactor operated by Consumers Power Company.

This yield varies by a factor of four and appears to be subject to other factors, as discussed in the previous paragraph. Table 1 also lists the fuel cladding material and the delay time between removal from the reactor and reprocessing. The data shows a greater yield of tritium as water vapor from the Zircaloy clad fuel than from the stainless steel clad fuel.

Dissolution samples

The results of the simultaneous gas and water vapor tritium samples obtained during three consecutive dissolutions of the Humboldt Bay campaign are shown in table 2. The burnup (MWD) for the three dissolution runs are sufficiently close for comparisons to be made between the three dissolution runs.

Table 2. Tritium in stack gas and water vapor for Humboldt Bay fuel batch numbers 7, 8, and 9

	Batch number[a]			Background
	7	8	9	
Total burnup (MW(t)-days)-------	9.7×10^{3}	9.8×10^{3}	8.8×10^{3}	0
Sampling time (hours)----------	8.3	8.0	19.4	7.0
Chemical form				
Water vapor concentration ($\mu Ci/cm^3$ of air)---	1.6×10^{-7}	2.2×10^{-7}	2.2×10^{-7}	1.1×10^{-7}
Total activity as water vapor (curies)----------	$.9 \times 10^{-1}$	1.2×10^{-1}	2.8×10^{-1}	5.2×10^{-2}
Gas concentration ($\mu Ci/cm^3$ of air)---	2.0×10^{-6}	[b] 1.9×10^{-6}	$.9 \times 10^{-6}$	4.0×10^{-9}
Total activity as gas (curies)-------	$1.1 \times 10^{+0}$	[b] $1.0 \times 10^{+0}$	$1.2 \times 10^{+0}$	1.9×10^{-3}

[a] The combined analytical and sampling 1σ errors are less than 12 percent except where noted by (b) the errors are less than 22 percent of the stated value.

The dissolution cycle is defined as the period of time starting when the chopped fuel is immersed in the acid bath and ending when the dissolved acid mix is transferred from the dissolving container. This time period varies from 18 to 48 hours and is generally around 24 hours. Sampling commenced at the beginning of the dissolution cycle and lasted 8 hours for the dissolution batches 7 and 8, and 20 hours for dissolution batch 9. The tritium stack background sample was obtained during a non-dissolution period, but fuel chopping operations were carried out during the background sampling run.

The tritium water vapor concentration measured during the three dissolutions averaged 2.0×10^{-7} $\mu Ci/cm^3$. The evolution rate of tritium as water vapor is fairly constant throughout the dissolution cycle as shown by comparing the 8-hour runs with the 20-hour run in terms of curies per hour.

Batch 7 (8 hours)---------- 1.1×10^{-2} Ci/h
Batch 8 (8 hours)---------- 1.5×10^{-2} Ci/h
Batch 9 (20 hours)---------- 1.4×10^{-2} Ci/h

The rate of release of the tritium from the fuel is not well defined but is assumed to occur during the early stage of the dissolution cycle. Once released from the fuel, the tritium as water becomes part of the dissolver solution. The evaporation of water vapor from the dissolver solution into the off-gas line is dependent upon the temperature of the solution. Since the temperature of the solution is maintained at a constant level throughout the dissolution cycle, the evaporation rate of the tritium as vapor is also constant.

The concentration of gaseous tritium measured during the three dissolutions of the Humboldt Bay campaign averaged 2.0×10^{-6} $\mu Ci/cm^3$ air for the 8-hour runs and 9.0×10^{-7} for the 20-hour run. The variation in concentration between the 8- and 20-hour observations indicates that the evolution of the gaseous tritium is not constant throughout the dissolution cycle. The larger fraction of the gaseous tritium is released from the fuel within the first 8 hours of the cycle. Unlike the tritium in the vapor form, the gaseous tritium does not become part of the dissolver solution but goes directly to the off-gas system. Therefore, the tritium release in the gaseous form was related to fuel burnup of the dissolution batch since its release is not a function of time.

Batch 7 (8 hours)---------- 1.1×10^{-4} Ci/MWD
Batch 8 (8 hours)---------- 1.0×10^{-4} Ci/MWD
Batch 9 (20 hours)---------- 1.3×10^{-4} Ci/MWD

Tank samples were obtained during the dissolution of fuel batches 7 and 8 to determine the gaseous tritium evolution rate. The sampling consisted of constant low flow-rate withdrawal into evacuated tanks for periods of approximately 3.7 hours. Two samples were obtained during each dissolution. The results of these samples are shown in table 3. Sampling commenced when dissolution started and the sampling times relative to dissolution startup are given. The concentrations obtained were used to project a normalized output over the sampling period. For batches 7 and 8, approximately 80 percent of the total activity output of gaseous tritium occurred during the first one and one-half hours of the sampling period. This type of output is similar to the krypton-85 gaseous output observed previously (6).

Campaign samples

Simultaneous water vapor and gaseous tritium samples were obtained during the Parr and Big Rock Point campaigns. These samples permitted a comparison to be made between the two tritium forms over an entire campaign period. The samples were continuous and yielded the results shown in table 4. The ratio of tritium gas to water vapor was 0.66 for the Parr campaign and 0.09 for the Big Rock Point campaign. The

Table 3. Tritium in gas evacuated tank samples, Humboldt Bay batch numbers 7 and 8

Dissolution batch	Sampling time relative to dissolution startup (hours)	Concentration ($\mu Ci/cm^3$ of air)	Percent of normalized stack output
7----------	0 - 1.6	1.15×10^{-5}	86
7----------	1.6 - 3.5	$.16 \times 10^{-5}$	14
8----------	0 - 1.4	1.31×10^{-5}	80
8----------	1.4 - 3.7	$.20 \times 10^{-5}$	20

Table 4. Tritium stack output[a] during campaign periods

Campaign	Total burnup (MW(t)-days)	Activity released (curies)			
		Vapor	Gas	Total	Per MWD
Parr - CVNPA-----	34,408	15.6	10.3	25.9	7.5×10^{-4}
Big Rock Point---	80,483	63.2	5.4	68.6	8.5×10^{-4}

[a]Based on composite samples over the entire campaign period. Sampling and analytical 1σ errors < 15 percent.

reason for this variation is unknown. It is interesting to note that even though there is a large variation in the gas to water vapor ratio, the total tritium yield per MWD for each campaign is of the same magnitude.

Tritium inventory

The samples collected during the Humboldt Bay campaign enable a projection to be made of the total tritium inventory for this campaign. The previously mentioned gaseous stack samples obtained during three dissolution cycles and the weekly water vapor samples obtained during the final 5 weeks of the campaign are the basis for the projected airborne tritium inventory. The liquid waste composite sample collected throughout the entire campaign enabled projection of tritium inventory of the liquid waste effluent. Some of the tritium remains in the stored high-level liquid waste. This is assumed to be 5 percent of the total tritium available in the fuel at reprocessing based upon volume ratio (8).

Table 5 shows the Humboldt Bay campaign fuel inventory in terms of curies per megawatt-days. Based upon total tritium yielded from the fuel, the stack tritium contribution is 7 percent while the liquid effluent contributes 88 percent. These values differ from the safety report projection of 25 percent to the stack and 65 percent to the liquid effluent (9).

The last column of table 5 shows the tritium inventory as a percent of the total tritium produced in the reactor per megawatt-day (1×10^{-2} Ci/MWD) (9). The inventory shows a deficit of 40 percent

Table 5. Tritium inventory for Humboldt Bay campaign

Source	Ci/MWD	Percent of total from fuel	Percent of predicted[a]
Liquid effluent	5.33×10^{-3}	88	53
High-level liquid	[b]3.18×10^{-4}	5	3
Stack vapor	3.09×10^{-4}	5	3
Stack gas	1.18×10^{-4}	2	1
Assumed loss	3.93×10^{-3}	-	40

[a]Based upon a total yield of 1×10^{-2} Ci/MWD for 20,000 MWD/tonne burnup (9).

[b]Assumed based upon plant records (8).

which was not accounted for during reprocessing. The deficit consists primarily of tritium which diffused through the cladding while the fuel was in the reactor and then stored prior to reprocessing. Some of the deficit can also be contributed to tritium off-gassed from the nitric acid recycle storage tank, buildup in the uranium product, and hydride associated with the cladding.

Discharge estimates

The in-plant data have been summarized in table 6. The data presented give actual measured values for stack releases of tritium in both the gaseous and vapor forms for four campaigns considered representative of typical fuel types normally processed at this plant.

Based upon the data in table 6, the average tritium stack output is 6.75×10^{-4} Ci/MWD which consists of 66 percent in the water vapor state and 24 percent in the gaseous state.

The total fuel burnup processed by NFS over the past 6 years is 4.1×10^{6} MWD, or 6.8×10^{5} MWD/year (8,10). The projected tritium stack output for the same periods are 2.8×10^{3} curies total or 4.6×10^{2} Ci/year based on the Ci/MWD factor derived from table 6.

Likewise, using the data derived from the Humboldt Bay interception sample (table 5), the liquid discharge rate of tritium amounts to 5.33×10^{-3} Ci/MWD. Using this discharge rate, the total liquid discharge of the plant for 6 years of operation is 2.2×10^{4} curies or 3.6×10^{3} Ci/year. This value correlates closely with present projections of the liquid tritium plant discharge (10).

Table 6. Summary, stack tritium effluent results

Campaign	Total burnup (MW(t)-days)	Gas (Ci)	Vapor (Ci)	Total (Ci)	Gas (Ci/MWD)	Vapor (Ci/MWD)	Total (Ci/MWD)
Humboldt Bay--	151,328	[a]17.7	46.8	64.5	1.2×10^{-4}	3.1×10^{-4}	4.3×10^{-4}
Yankee Rowe---	224,730	-	45.8	-	-	2.0×10^{-4}	-
Parr----------	34,308	10.3	15.6	25.9	3.0×10^{-4}	4.5×10^{-4}	7.5×10^{-4}
Big Rock Point	80,483	5.4	63.2	68.6	$.6 \times 10^{-4}$	7.9×10^{-4}	8.6×10^{-4}

[a]Based upon three dissolution cycles. All other values based on measurements throughout the entire campaign periods.

Field sampling results

The tritium field samplers were placed at five locations around the plant boundary. The exact location of each sampler is shown in figure 1. Weekly samples were obtained during June 5 to October 4, 1971. During this time period, the plant processed a portion of the Humboldt Bay fuel and the entire Yankee Rowe fuel campaign. The weekly results of all tritium concentrations from each field station are shown in table 7. Station numbers 1 through 5 correspond to the locations shown in figure 1 and station number 6 is the control station located at Winchester, Mass.

The air concentrations of tritium observed at each station represent background levels plus plant-related contributions. Plant contributions include evaporation from the liquid waste lagoon system, receiving streams, and solid waste disposal area as well as stack discharge. On a weekly basis, the field stations do not show any significant contributions of airborne tritium that can be related to the plant. The plant contribution is not observed because it is small compared to the normal week-to-week fluctuations of the background tritium.

The weekly data have been compared with weekly wind-rose data (11) in order to determine if a trend of concentration variation could be associated with the general wind patterns at the plant site but no correlation can be demonstrated. Relative humidity effects are minimal. Some dilution is evident at high humidity levels but the air tritium concentration remains the same.

The average tritium concentrations for the entire study period at each station are shown in table 8. These average values are derived by compositing the weekly concentrations. Values are given both in terms of activity per volume of water vapor sampled and activity per volume of air sampled. In general, the levels observed at the five stations are within the background range observed at the control station. The ratio of air concentration to water vapor concentration is constant for all stations including the control, with a mean value of 1.2×10^{-5} $\mu Ci/cm^3$ air per $\mu Ci/cm^3$ water.

The average air concentrations of tritium in terms of activity per volume of water vapor can be compared with observations of Daly et al. (5) made at equivalent locations around NFS during plant operation in 1967. Daly's data consists of 3-day samples using bulk desiccant at multiple stations. Table 9 is a presentation of the average levels of tritium observed from July 17 to September 23, 1967. Distance from the plant is stated as a range since multiple stations were used in each direction shown. Daly's results are very close to our 1971 values. Both the 1971 and 1967 data indicate higher average tritium concentrations northwest of the plant when compared to the other sample locations.

Table 7. Weekly air concentrations of tritium around NFS during 1971

Week of:	Concentration[a] 10^{-5} pCi/cm^3					
	Station 1	Station 2	Station 3	Station 4	Station 5	Station 6 (control)
June 7	4.3	2.0	1.2	1.9	0.9	0.9
June 14	1.9	1.8	2.3	1.5	4.4	9.2
June 21	2.2	2.2	2.1	1.8	8.1	6.6
June 28	3.4	3.2	2.4	2.0	5.9	6.7
July 5	2.6	2.8	1.2	NS	5.3	2.4
July 12	2.2	1.9	1.4	NS	4.1	2.2
July 19	1.9	2.2	1.6	NS	7.8	2.1
July 26	2.9	1.8	2.6	NS	3.4	1.8
August 2	2.3	1.9	1.9	3.1	3.0	1.3
August 9	2.2	2.4	2.5	2.0	4.1	.8
August 16	NS	2.6	2.7	1.8	4.3	1.3
August 23	NS	1.5	2.0	1.0	3.6	1.6
August 30	1.8	1.4	1.6	2.1	3.7	1.6
September 6	2.3	1.6	1.3	8.6	6.7	.4
September 13	.7	.6	1.0	.9	2.9	.6
September 20	.8	.7	.7	.6	1.7	.4
September 27	1.6	1.0	1.0	1.0	4.1	.9

[a]The combined analytical and sampling 1σ errors are < 15 percent.
NS, no sample collected.

Table 8. Average tritium in air concentrations around NFS during summer of 1971

Station (EPA no.)	Direction from plant	Distance (km)	Average concentration		Concentration ratio of air to water
			$\left(\frac{pCi}{cm^3\ H_2O}\right)$	$\left(\frac{pCi}{cm^3\ air}\right)$	
1----------	NNE	3.4	1.9×10^{0}	2.2×10^{-5}	1.2×10^{-5}
2----------	NE	2.0	1.8×10^{0}	1.9×10^{-5}	1.1×10^{-5}
3----------	SE	2.8	1.7×10^{0}	2.1×10^{-5}	1.2×10^{-5}
4----------	SW	2.4	1.3×10^{0}	1.6×10^{-5}	1.2×10^{-5}
5----------	NW	2.8	3.3×10^{0}	4.4×10^{-5}	1.3×10^{-5}
6----------	Control	(a)	2.4×10^{0}	2.4×10^{-5}	1.0×10^{-5}

[a]Control located at Winchester, Mass.

Table 9. Specific activity of airborne water vapor at NFS during summer of 1967 (5)

Station	Direction	Distance (km)	Average specific activity ($pCi/cm^3\ H_2O$)
Daly A----------	N	2.4 - 6.4	2.3 x 10
B----------	NE	2.0 - 5.3	1.8 x 10
D----------	SE	2.4 - 5.5	1.7 x 10
F----------	SW	.8 - 6.4	1.4 x 10
G----------	NW	.8 - 4.5	5.3 x 10

Further evidence of elevated tritium in this direction is available from produce composite samples (12) obtained at five locations around the NFS plant, corresponding to the five tritium air sampling stations. The total tritium concentrations from the farm vegetable composites are shown in table 10. The farm sample northwest of the plant shows an elevated tritium concentration relative to the other four locations. The tritium level northwest of the plant cannot be explained in specific terms because necessary detailed pathway data are not available to describe this phenomena. However, we can assume that the northwest direction is influenced somewhat by evaporation from the Buttermilk Creek, the main pathway for tritium discharged as liquid waste from the NFS lagoon system, and beyond this we can only speculate on the micrometeorology in this sector that possibly influences this phenomena.

Dose estimates

The tritium data obtained from this study enables the projection of dose directly from the field measurements as well as projecting the contribution to dose from the stack tritium discharge.

The dose calculations are based upon the International Commission on Radiation Protection (ICRP) recommendations (13) with a correction factor of 1.5 to account for the increase in dose due to organic labeling from chronic exposure (14). A quality factor of 1.0 for tritium is assumed in these calculations (15).

Table 10. Average tritium in produce composites around NFS during summer of 1970

Station	Direction	Distance from plant (km)	Tritium concentration[a] (pCi/g wet weight)
Farm 6------	NNE	3.6	0.6 ± 0.3
Farm 5------	NE	2.0	.6 ± .3
Farm 3------	SE	2.8	.7 ± .3
Farm 2------	SW	2.4	.4 ± .2
Farm 1------	NW	3.0	1.1 ± .4
Control-----	NE	24	.5 ± .3

[a]Error is the 2σ error due to counting statistics.

Total body dose = 1.7×10^3 (X) μrem/year

Where

X = average tritium air concentration (pCi/cm^3 air).

Table 11 shows the calculated whole body dose from tritium around NFS. The annual dose is based on the average tritium concentrations observed at each station location (table 8) projected to an annual average. The stack dose contribution is based upon the estimated annual stack output of 460 curies of tritium per year and is projected to the property line in the direction of each sampling station using current wind-rose data obtained at the NFS stack (11). (See appendix B-2.) These dose estimates show that the NFS stack contribution was 0.5 percent to 3 percent of the total dose from environmental tritium.

It is not possible from the present study to ascertain the airborne environmental dose contribution from evaporation of the liquid tritium effluent.

Table 11. Whole body dose from airborne tritium (microrem per year)

Station	Direction	From all sources	NFS stack contribution
1----------	NNE	60	1.5
2----------	NE	50	1.5
3----------	SE	60	.3
4----------	SW	40	.3
5----------	NW	110	.8
6----------	Control	60	-

CONCLUSIONS

Tritium in airborne effluents consists of both the water vapor and the gaseous form. The water vapor form is discharged continuously during a campaign period, and originates from dissolution and various evaporative processes ongoing during a campaign. The discharge rate of the water vapor tritium is 5×10^{-4} Ci/MWD of fuel processed. The

gaseous tritium is primarily discharged during the initial dissolution of fuel although a small amount is discharged during the fuel chopping operation. The discharge rate of the gaseous tritium is approximately 2×10^{-4} Ci/MWD. Based upon a reprocessing production schedule of 6.8×10^{5} MWD per year, the total tritium stack discharge rate is approximately 15 μCi/s.

The impact of tritium as stack discharge is minimal when compared to the normal environmental level of airborne tritium. Based upon measurements from this study, the average tritium background around NFS is approximately 2.4×10^{-5} pCi/cm^3 air and the stack contribution is estimated to be from 0.5 to 3 percent of the background tritium concentration.

In terms of dose, the stack discharge contributes a total body dose of less than 2 μrem annually outside the plant boundary. The air concentration from evaporation of liquid discharges has not been estimated but is assumed to be less than the stack contribution with the possible exception of the sector northwest of the plant. In this sector, due to the close proximity of the Buttermilk Creek, evaporation from the creek may contribute to the elevated tritium air concentrations observed. This cannot be confirmed from the present study since the levels in the sector, although elevated, are still within the background range.

The samplers used during this study proved to be both dependable and adequate for the designed purpose of sampling tritium in both the vapor and gaseous states. The desiccant approach to the collection of water vapor is straight-forward and simple, which is advantageous under field conditions. The combustion-desiccant device used for the collection of both gas and vapor components of tritium in the stack proved dependable over long periods of sampling (30 to 40 days) which permitted long-term samples to be collected and also permitted continuous sampling of the stack effluent over an entire campaign period.

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APPENDIXES

APPENDIX A. SAMPLER DESCRIPTION

Tritium sampler description

The following two sections of this appendix describe the tritium samplers used both at the stack and in the environment during the tritium study at NFS.

A more detailed discussion of the sampler and its operation, including the laboratory testing and calibration will be found in reference (7), "A Sampler for Non-Aqueous Tritium Gases" by Griffin, et al.

Stack tritium sampler

The sampler consists of the following items:

(a) Two desiccant traps each with a capacity of 60 grams of water. The desiccant consists of approximately 1,000 grams of anhydrous calcium sulphate, 8 mesh (Hammond Drierite Company).
(b) One catalytic burner consisting of a trap containing 0.5 percent platinum on aluminum oxide pellets (Englehard Industries, Inc.). The trap is inserted into a heating jacket and the system operates at 400° C.
(c) One tank of 2 percent hydrogen in air-carrier gas.
(d) Various flow meters, manometers, valves, pump, and other hardware.

The system samples filtered stack effluent at a flow rate of 0.2 liters per minute. The stack gas initially passes through a flow meter and then into the first desiccant. Moisture is removed in this trap to an equivalent -79° C. dewpoint (0.003 mg/liter). The dried stack gas existing from the desiccant trap is mixed with carrier hydrogen, and this mixture is injected into the catalytic burner.

The burner oxidizes the gaseous hydrogen compounds contained in the stack gas, as well as the carrier, to the water vapor state. The hot gases leave the burner and pass through a copper tubing coil which allows the gas to cool. The cooled gas passes through the second desiccant trap where water vapor produced in the burner is collected.

The stack gas and carrier gas flow rates can be varied so that adequate sample size can be obtained over short-term periods of several hours to periods of several weeks. In situations where only water vapor is to be sampled, the carrier gas, burner, and second desiccant trap are removed from the system.

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Water vapor collected in the desiccant traps is removed using a specially designed dehydration unit. The dehydration process is very efficient (97 percent recovery of activity) and yields a water sample which is then filtered and distilled to remove any suspended desiccant material and insure a neutral pH. The sample is analyzed using standard liquid scintillation technique.

Field atmospheric tritium sampler

The field sampler operates essentially as the first stage of the stack unit, sampling only water vapor. Unlike the stack sampler, this sampler operates over weekly periods under varying relative humidity conditions. To accommodate for this variability, three desiccant traps are utilized in series at a flow rate that allows for a reasonable sample during a dry week and the capability to collect a large sample without saturating the total trap capacity during a wet week.

The sampler consists of the following:

(a) three desiccant traps each with a capacity of 60 grams of water,
(b) a flow meter, and
(c) an air pump.

During any given sampling period, only traps that contain sampled water vapor are dehydrated and analyzed. The remaining traps are reused during the following sampling period.

APPENDIX B. PLANT DATA

Table B-1. Campaign data for fuel inventories processed during tritium study at NFS during 1971 (8)

Campaign	Total burnup (MW(t)-days)	Tonnes total	Fuel age (years)	Fuel cladding	Number of dissolutions	Processing period
Humboldt Bay (PGE)	151,328	21	3.53	Zr, SS-304	23	4/25 7/02
Yankee Rowe	224,730	10	.77	SS	14	7/12 8/24
Parr (CVNPA)	34,308	3.4	4.80	Zr-4	10	9/21 10/04
Big Rock Point	80,438	5.9	1.58	Zr-Z, Iconel	7	10/10 11/08

Table B-2. Average X/Q values for the NFS site during 1971[a] (11)

Wind direction	Boundary distance (meters)	Average X/Q[b] (s/m^3)	
		Summer	Annual
N	2,200	7.08×10^{-9}	1.27×10^{-8}
NNE	2,000	6.60×10^{-9}	8.98×10^{-9}
NE	2,000	6.32×10^{-9}	7.12×10^{-9}
ENE	1,600	6.18×10^{-9}	1.59×10^{-8}
E	1,400	8.44×10^{-9}	1.41×10^{-8}
ESE	1,200	1.21×10^{-8}	1.47×10^{-8}
SE	1,200	3.86×10^{-8}	2.21×10^{-8}
SSE	3,500	2.56×10^{-8}	1.18×10^{-8}
S	2,400	2.19×10^{-8}	1.63×10^{-8}
SSW	1,600	3.99×10^{-8}	4.10×10^{-8}
SW	1,600	3.87×10^{-8}	3.91×10^{-8}
WSW	1,800	1.95×10^{-8}	2.84×10^{-8}
W	2,400	1.40×10^{-8}	1.50×10^{-8}
WNW	2,600	1.62×10^{-8}	1.06×10^{-8}
NW	2,800	1.01×10^{-8}	8.66×10^{-9}
NNW	2,400	9.13×10^{-9}	1.01×10^{-8}

[a]Based upon hourly averages obtained from instrumentation located at the top of the stack.

[b]X/Q = air concentration in Ci/m^3 per Ci/s release.

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THE ABSTRACT CARDS accompanying this report are designed to facilitate information retrieval. They provide suggested key words, bibliographic information, and an abstract. The key word concept of reference material filing is readily adaptable to a variety of filing systems ranging from manual-visual to electronic data processing. The cards are furnished in triplicate to allow for flexibility in their use.

OBSERVATION OF AIRBORNE TRITIUM WASTE DISCHARGE FROM A NUCLEAR FUEL REPROCESSING PLANT, EPA/ORP 73-1. J. A. Cochran, W. R. Griffin, Jr., and E. J. Troianello (February 1973).

ABSTRACT: A study was conducted at Nuclear Fuel Services, Inc. (NFS) to: (a) characterize the stack tritium effluent in the gaseous and water vapor forms, during normal plant operations, (b) determine the weekly average tritium air concentrations at five selected sites around the plant over a 4-month period, (c) evaluate methods used for sampling at the stack and in the environment, and (d) estimate the dose to the population in the immediate vicinity of the plant from tritium stack effluents.

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(over)

Characterization of tritium release from the stack was carried out during three reprocessing campaigns assumed to be typical of the NFS operation. Evaluation of the data indicates that the tritium release rate from the stack is 6.75×10^{-4} curies per megawatt-day burnup. Approximately 25 percent of the activity is in the gaseous state and 75 percent is in the water vapor state. The average annual release of tritium via the stack, considering the total fuel processed from 1966 to 1971, is estimated to be 500 curies per year. Data for the five air sampling stations around the site indicate that the plant contribution to the atmosphere is small.

KEY WORDS: Analysis; fuel reprocessing; nuclear; population dose; sampling; tritium; waste discharge.

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